# UTILITY PATENT APPLICATION TRANSMITTAL

(Only for new nonprovisional applications under 37 CFR 1.53(b))

| | |
|---|---|
| *Title of Invention* | **Biological De-Inking Method** |
| *Named Inventor(s)* | **Tae Jin Eom**<br>**Steven Say-Kyoun Ow** |
| *Attorney Docket* | **20565-0111** |
| *Express Mail Label No.* | **EM 515 354 958 US** |

jc503 U.S. PTO 07/22/98

jc135 U.S. PTO 09/121152 07/22/98

## APPLICATION ELEMENTS

1. [X] Fee Transmittal Form
*(Submit an original, and a duplicate for fee processing)*
2. [X] Specification, Claims, and Abstract — Total Pages **18**
3. [ ] Drawings — Total Sheets ____
4. Oath or Declaration — Total Pages _____
   a. [ ] Newly executed (original or copy)
   b. [X] Copy from prior application (37 CFR 1.63(d)) *(for continuation/divisional with Box 17 completed)*
   **[Note Box 5 Below]**
   (i) [ ] DELETION OF INVENTOR(S)
   Signed statement attached deleting inventor(s) named in the prior application, see 37 CFR 1.63(d)(2) and 1.33(b).
5. [X] Incorporation by Reference *(usable if Box 4b is checked)*
The entire disclosure of the prior application, from which a copy of the oath or declaration is supplied under Box 4b, is considered as being part of the disclosure of the accompanying application and is hereby incorporated by reference therein.
6. [ ] Microfiche Computer Program *(Appendix)*
7. [ ] Nucleotide and/or Amino Acid Sequence Submission *(if applicable, all necessary)*
   a. [ ] Computer Readable Copy
   b. [ ] Paper Copy (identical to computer copy)
   c. [ ] Statement verifying identity of above copies

*ADDRESS TO:* Assistant Commissioner for Patents
Box Patent Application
Washington, D.C. 20231

## ACCOMPANYING APPLICATION PARTS

8. [X] The Assignment Papers can be found in the prior application, assigned to Korea Research Institute of Chemical Technology
9. [ ] 37 CFR 3.73(b) Statement *(when there is an assignee)*
   [ ] Power of Attorney by assignee
10. [ ] English Translation Document *(if applicable)*
11. [ ] Information Disclosure Statement (IDS) PTO-1449
   [ ] Copies of IDS Citations
12. [X] Preliminary Amendment
13. [X] Return Receipt Postcard (MPEP 503) *(Should be specifically itemized)*
14. [X] Small Entity Statement(s)
   [X] Statement filed in prior application Status still proper and desired
15. [ ] Certified Copy of Priority Document(s)
16. [ ] Other: _____

17. If a **CONTINUING APPLICATION**, *check appropriate box and supply the requisite information*:
[X] Continuation of prior application No: 08/239,313, filed May 6, 1994

18. CORRESPONDENCE ADDRESS:

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By: _____ Reg. No.36,714
Date: July 22, 1998
Telephone: 404-818-3700
Facsimile: 404-818-3799

Patents
20565-0111

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

In re Application of:

**TAE JIN EOM et al.**

Serial No. **Not Yet Assigned**

Filed: **July 22, 1998**

For: **BIOLOGICAL DE-INKING METHOD**

## PRELIMINARY AMENDMENT

Assistant Commissioner for Patents
Washington, DC 20231

Sir:

Prior to examination of the patent application identified above, please enter the following Preliminary Amendment and consider the following remarks.

**In the Specification**

Please amend the Specification as follows:

On page 6, line 3, please delete "consistency 2-15%" and replace therefor --consistency 12-15%--.

On page 6, line 6, please delete the phrase "adjusted in the acidic range, i.e.," and replace therefor --in the range of 3.0 to 8.0, preferably--.

**In the Claims**

Please cancel Claim 1-20 without prejudice or disclaimer.

Please add the following new claims:

**Express Mail Mailing Label No. EM 515 354 958 US**

**Date of Deposit: July 22, 1998**

--21. A method of de-inking waste printed paper, comprising

a) pulping waste printed paper with an enzyme capable of dislodging ink particles from the waste printed paper in an aqueous medium at a pH between about 3 to 8, and wherein the ink is dislodged from the waste printed paper by action of said enzyme; and

b) removing dislodged ink particles from the resulting pulp containing medium.

22. The method of Claim 21 wherein dislodged ink particles are removed by flotation.

23. The method of Claim 21 wherein dislodged ink particles are removed by washing.

24. The method of Claim 21 wherein the amount of enzyme used is in the range of about 0.005 to about 5 percent-by-weight based on the dry weight of the wastepaper.

25. The method of Claim 21 wherein said enzyme is selected from the class consisting of cellulase, hemicellulase, pectinase, other carbohydrases and mixtures thereof.

26. The method of Claim 21 wherein said enzyme is a cellulase selected from the group consisting of cellulases derived from *Trichoderma viride, Aspergillus niger* and mixtures thereof.

27. The method of Claim 21 wherein the pH of said aqueous medium is from about 3 to 7.

28. The method of Claim 21 wherein the pulping occurs at a consistency of pulp of about 12% or greater.

29. The method of Claim 21 wherein the pulping occurs for a period of less than about 1 hour.

30. The method of Claim 21 wherein the temperature of the pulping is in a range of from about 20°C up to about 60°C.

31. A method of recycling waste printed paper, comprising

a) pulping waste printed paper;

b) contacting waste printed paper with an enzyme capable of dislodging ink particles from the waste printed paper in an aqueous medium at a pH between about 3 to 8, and wherein the ink is dislodged from the waste printed paper by action of said enzyme; and

c) removing dislodged ink particles from the resulting pulp containing medium.

32. The method of Claim 31, wherein the enzyme is a cellulase selected from the group of cellulases derived from *Trichoderma viride, Aspergillus niger* or mixtures thereof wherein said cellulase is used in an amount between about 0.005 and about 5.0 percent-by-weight based on the dry weight of said waste printed paper, said contacting being carried out at a temperature between about 20°C and about 60°C.

33. The method of Claim 31 wherein the amount of enzyme used is in the range of about 0.005 to about 5.0 percent-by-weight based on the dry weight of the wastepaper.

34. The method of Claim 31 wherein the enzyme is selected from the class consisting of cellulase, hemicellulase, pectinase, other carbohydrases and mixtures thereof.

35. The method of Claim 31 wherein said enzyme is a cellulase selected from the group consisting of cellulases derived from *Trichoderma viride, Aspergillus niger* and mixtures thereof.

36. The method of Claim 31 wherein the ink particles are removed by flotation or washing.

37. The method of Claim 31 wherein the pH of said aqueous medium is from about 3 to 7.

38. The method of Claim 31 wherein the pulping occurs at a consistency of pulp of about 12% or greater.

39. The method of Claim 31 wherein the pulping occurs for a period of less than about 1 hour.

40. The method of Claim 31 wherein the temperature of the pulping is in a range of from about 20°C up to about 60°C.

41. A method of biologically de-inking waste printed paper comprising;

a) pulping the waste printed paper with an enzyme capable of dislodging ink particles from the waste printed paper in an aqueous medium at an acidic range or neutral range pH, and wherein the ink is dislodged from the waste printed paper by action of said enzyme; and

b) removing dislodged ink particles from the resulting pulp containing medium.--

## REMARKS

Claims 21-41 are now pending in the above-identified patent application as of this Preliminary Amendment. No new matter is contained in the amendments. Support for the pH range of from 3 to 8 added in the current Specification and in Claims 21 and 31, can be found, for example, on page 6, line 8, of parent application Serial No. 07/518,935. Support for the language regarding high consistency pulping consistency of 12% or greater added in the current Specification and in Claims 28 and 38, can be found, for example, on page 6, lines 1-4, of parent application Serial No. 07/518,935. Support for the language regarding pulping for less than about 1 hour in Claims 29 and 39 can be found, for example, in each of the Examples of the present Specification. Support for the language regarding temperature of the pulping in Claims 30 and 40 can be found, for example, in former Claims 8 and 32.

All de-inking prior to the time of Applicants' invention was performed in an alkaline environment, as it was then believed that alkali was needed to swell the fibers to detach ink from fiber surface. Applicants' invention is a novel and unobvious improvement, which allows de-inking to be performed in an acidic or neutral medium. "Neutral medium" refers to a fibrous suspension of disintegrated waste paper, the pH of which has not been modified by the deliberate addition of chemical agents in order to give it a substantially alkaline character. Depending upon the origins of the water and paper used, the pH of this neutral medium can be above 8, given that the minerals in the water or fillers and binders in the paper can impart a slight alkalinity to the suspension.

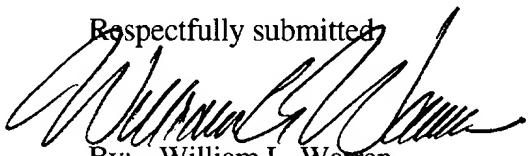
Applicants have entered into the claims the limitation that the pH of the deinking medium is from about 3 to 8, or is in the acidic range or neutral range pH. Applicants intend for the claims to exclude the practice of adding alkali for the purpose of achieving the prior art alkaline

deinking technique. The claims should not be construed to exclude the case where an infringer adds an immaterial amount of alkali for the purpose of attempting to avoid infringement, while achieving an equivalent of the claimed invention by the same function, way and result.

No additional fees are believed due; however, the Commissioner is hereby authorized to charge any additional fees which may be required or credit any overpayment to deposit account no. 10-1215.

This preliminary Amendment places all claims in the present application in condition for allowance, and such action is courteously solicited. The Examiner is invited and encouraged to contact the undersigned attorney of record if such contact will facilitate an efficient examination and allowance of the application.

Respectfully submitted,



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Attorney Docket No.: 20565-0111

SPECIFICATION

Docket No. P673

TO ALL WHOM IT MAY CONCERN:

BE IT KNOWN, that We, Tae Jin EOM, a citizen of Korea residing in Daejonjikhal-shi, Korea, and Steven Say-kyoun OW, a citizen of the United States of America residing in Daejonkikhal-shi, Korea, have invented new and useful improvements in a

BIOLOGICAL DE-INKING METHOD

of which the following is a specification:

**CERTIFICATE OF MAILING**

I hereby certify that this correspondence and all referenced enclosures are being deposited with the United States Postal Service as Express Mail No. TB271329146 in an envelope addressed to: Commissioner of Patents and Trademarks, Washington, DC 20231 on May 6, 1994.

Pamela Riggs

## Background of the Invention

This is a continuation-in-part application of United States Patent Application Serial No. 07/518,935, filed May 4, 1990, co-pending herewith and later abandoned.

### 1. Field of the Invention

This invention relates to a process for reclaiming useful pulp fibers from wood-containing or wood-free wastepaper by a biological method in the de-inking process.

### 2. Description of the Background

De-inking of pulp fibers is essentially a laundering or cleaning process in which the ink is considered to be the dirt.

Chemicals, along with heat and mechanical energy, are used to dislodge the ink particles from fibers and to disperse them in the aqueous medium. The ink particles are then separated from the pulp fibers, either by washing or flotation or by using a modern hybrid process that combines washing and flotation.

The chemicals used for the conventional de-inking process are surfactants which function as detergents to remove ink from the fiber, as dispersants to keep the ink particles dispersed and prevent redeposition on the fibers, and foaming agents in the froth flotation of ink particles.

A typical surfactant is a long chain molecule with a hydrophobic part on one end and a hydrophilic part on the other end. The hydrophobic part may consist of fatty acid, fatty alcohol, alkylphenols or other oil-soluble surfactants.

The hydrophilic part in the de-inking surfactant usually consists of anionic molecules, such as carboxylic acid salts or sulfonic acid salts and nonionic molecules, such as polyoxyethylenated chains.

The typical surfactants commonly used in the washing and froth flotation de-inking processes are: sodium and potassium salts of straight-chain fatty acids (soaps), linear alkylbenzenesulfonates (LAS), long-chain fatty alcohols, polyoxyethoxylated alkylphenols, alkyiphenol ethoxylates, and polyoxyethoxylated straight-chain alcohols.

A major disadvantage of using these surfactants in the de-inking process is excess foaming in the subsequent pulp stock flow and paper making process lines. Moreover, some of the above surfactants are resistant to biodegradation in the effluent treatment stages causing a serious environmental problem.

In the froth flotation de-inking process, collector is added to agglomerate ink into large particles and attach them to the air bubbles. Collectors are required for effective flotation and are usually anionic long-chain fatty acid soaps. Fatty acid collectors are precipitated with calcium ions to form larger, insoluble ink particles and collector particles. With injection of air in the flotation cells, the agglomerated ink particles adhere to the bubbles, rise to the surface and are skimmed off from the system.

Major disadvantages of the flotation method using the fatty acid collector are a pitch deposition and calcium scaling problems in the subsequent stock lines and papermaking process equipment. Besides surfactants, other chemicals employed are caustic soda, sodium silicate, metal ion chelating agents and hydrogen peroxide.

The hydrogen peroxide bleaching agent has to be added in order to prevent pulp yellowing caused by the addition of caustic soda and to improve the brightness of pulp fibers.

With advances in modern printing and photocopying technology, conventional de-inking with the aid of surfactants encounters serious problems because the wastepaper is printed with the use of heavily coated, highly polymerized or non-impact inks, such as ultraviolet, heatset, Xerox, laser and ink jet. These inks usually contain cured polymer

resins which bind ink particles so strongly on the fiber surface that it is impossible to dislodge the inks completely during the wastepaper defiberizing (pulping) stage with the conventional de-inking chemicals. Excess heat and mechanical energy are also required along with the ineffective conventional chemicals.

Furthermore, in the conventional flotation de-inking process for newsprint wastepaper, a major technical problem has to do with the fact that fine ink particles are embedded in the fiber bundles and between fibrils which are almost impossible to be removed from the fibers by a washing and/or flotation process.

## Summary of the Invention

This invention provides a new and much improved de-inking method which is effective for newsprint as well as wood-free printed wastepaper, such as whiteledger, laser printed, xerographic copypaper and computer printout wastepaper.

The de-inking method of the present invention is to remove ink particles by the use of the biological activity of enzymes on the cellulose fiber surface and the dispersing function of enzyme protein on ink particles.

In contrast to the conventional method, no alkali or de-inking surfactants are required. In the froth flotation process, the fatty acid collectors are not required. Since caustic soda is not used in the newsprint de-inking, a hydrogen peroxide bleaching agent is also not required for preventing yellowing .

The elimination of the fatty acid collector in this biological de-inking process will solve the persistent pitch and scale deposition problem associated with the conventional flotation process using the fatty acid type soap and calcium salts and silicates.

## Description of the Preferred Embodiment

The wastepaper, such as old newsprint, is disintegrated in a conventional pulper (consistency 4-7%) or in a high consistency pulper (consistency 2-15%) at a water temperature ranging from about 20°C up to 60°C. The addition level of enzyme is about 0.005% to about 5.0% by weight based on dry weight of wastepaper. The pH of the stock slurry is adjusted in the acidic range, i.e., below about 7.0, and preferably between about of 3.0 and 7.0.

As compared to the conventional pulping process using caustic and surfactants, the pulping in the presence of enzyme can be completed in a relatively short period and ink particles are completely separated from the fiber surface and dispersed well. The dispersed inks are removed out of the pulp fibers by using the conventional washing process equipment, such as vibrating screen and drum washers without the aid of detergent surfactants in single and multi-stages. The ink particles dispersed with the action of enzyme protein can be also selectively removed out of the diluted pulp slurry with conventional flotation equipment in which air is injected or drawn into the pulp to provide bubbles to pick up the particles. No fatty acid collector is required in the case of waste newsprint. A small amount of fatty acid collector may be added to enhance the ink removal efficiency in the case of laser-printed wastepaper.

Among enzymes which can be used in the method are the carbohydrases and particular enzymes, such as cellulase, hemicellulase, pectinase and mixtures thereof. Examples of commercially available cellulases useful in these methods are those derived from *Trichoderma viride* and *Aspergillus niger*. These and other acid resistant enzymes may be used alone or as mixtures.

This biological de-inking process lowers pulping energy requirements to a large extent (almost 50% reduction) since the addition of enzyme results in a substantial reduction in pulping time as compared to the pulping in the absence of enzyme. The observed faster and easier pulping in the presence of enzyme may be attributed to the unique biological activity of the enzyme which is effective to debond the fiber bonding and dislodge the inks bonded on the fiber surface as well as within the fiber bundles or between the fibrils. A partial enzymatic hydrolysis of cellulose within the micro structure of the fiber surface may occur during the pulping stage. Because of this biological activity of the enzyme, the fine ink particles embedded within fiber bundles, fibrils and fines which have been impossible to be taken out by the conventional de-inking chemicals in the case of old newsprint de-inking are removed.

According to this biological de-inking method of old newsprint, the addition of hydrogen peroxide to prevent fiber yellowing is not required. This results in a substantial reduction of de-inking chemical cost as compared to the conventional de-inking process using caustic soda, hydrogen peroxide, chelating agents and sodium silicates.

It should be pointed out that the physical strength properties of the resulting pulp fiber prepared by this inventive method are found to be higher than those of the corresponding pulp prepared by the conventional method in addition to the much higher resulting pulp brightness. The enzyme addition does not appear to degrade the fiber strength, but rather improves the fiber strength for reasons not presently known.

To more fully illustrate the present invention, the following non-limiting examples are presented.

## De-inking of Old Newsprint with a Cellulolytic-Enzyme

### Example 1

A sample of old newsprint wastepaper was added to the pulper which was filled with 40°C water at a consistency of 4% and a cellulase (enzyme) was dissolved at the dosage level of 0.1% based on oven dry weight of wastepaper. The wastepaper was soaked for 10 minutes and then disintegrated for 5 minutes. After a complete disintegration of wastepaper, one half of the pulp slurry was diluted to 1% consistency.

The diluted pulp slurry was moved to the air flotation cell and then the dispersed ink particles were removed out of the pulp slurry by skimming off the ink particles froth out of the cell while injecting air through a porous plate. The flotation time for the complete removal of the ink froth was one minute.

The other half of the pulp slurry was washed on a laboratory vibrating screen to remove the dispersed ink particles.

The resulting recycled pulp fibers obtained by the flotation and the washing step were evaluated for pulp brightness and mechanical strength properties.

### Comparative Example

To compare the foregoing enzyme-treated de-inked pulp to the conventional de-inked pulp, the same sample of wastepaper was treated in the pulper with addition of 1.0% sodium hydroxide, 0.3% hydrogen peroxide, 3% sodium silicate solution (water glass) and 0.8% of SERFAX MT-90 ( fatty acid soap) and 0.2% IGEPAL-660 (biodegradable nonionic surfactant marketed by GAF Corporation) based on oven dry weight of wastepaper. The pulping time was 10 minutes for a complete disintegration. After diluting to 1% consistency, the dispersed ink particles were removed by the flotation method with the laboratory flotation cell as described above.

| Table 1 | Comparison of properties of recycled pulp by the method of the present invention and the conventional method | | | | | |
|---|---|---|---|---|---|---|
| | Brightness (%) | | Tensile Index (N.m/g) | | Tear Index (mN.m/g) | |
| | KONP | AONP | KONP | AONP | KONP | AONP |
| cellulase enzyme | 47.1 | 45.2 | 28.9 | 32.4 | 11.7 | 13.6 |
| after washing | 50.3 | 48.6 | 29.3 | 32.9 | 11.8 | 14.1 |
| conventional method | 45.1 | 38.4 | 30.1 | 32.8 | 10.8 | 13.1 |
| KONP: Korean old newspaper<br>AONP: American old newspaper | | | | | | |

As shown in Table 1, the brightness of the pulp de-inked with enzyme was much higher than that of the pulp de-inked with the conventional chemicals and the mechanical strength of the pulp de-inked with enzyme was also superior to the pulp de-inked with the fatty acid collector and the dispersant (IGEPAL-660). Microscopic observation revealed that the pulp prepared by the present invention contained more long fiber fractions, has smoother fiber surface and looks less mechanically damaged.

As can be seen from the data in Table 1, the enzyme treated pulp gave cleaner and brighter pulp with the washing as compared to the flotation ink removal using the conventional method.

**Enzyme Concentration Effect On Disintegration Time**

The enzyme addition also appeared to accelerate the wastepaper disintegration to a large extent. When the old newspaper was disintegrated in the conventional pulper at the 4% consistency, the addition of 0.5% enzyme reduced the pulping time from 5 minutes (no enzyme addition) to 30 seconds for a complete disintegration as shown in Table 2.

| Table 2 | Relation between enzyme addition and disintegration time | | |
|---|---|---|---|
| enzyme (%) (cellulase) | 0.5 | 0.1 | 0 |
| disintegration time | 30> | 60-120 | 300< |

**De-inking of Laser CPO (computer printout) with Cellulolytic Enzyme.**

It is almost impossible to achieve a complete removal of laser beam cured ink particles from the laser CPO wastepaper with conventional de-inking chemicals, because the ink particles are so strongly adhered to the fiber surface that alkali and general de-inking surfactants used in the conventional process are not able to dislodge and disperse the ink in the pulp-water slurry.

**Example 2**

A sample of laser CPO wastepaper was added to water in a laboratory high consistency pulper to achieve a consistency of 12.5% and a cellulase (enzyme) was added to the water at the dosage level of 0.2% based on the dry weight of paper. At stock water temperature of 20-35°C, the pulping was carried out for 20 minutes. The completely disintegrated pulp slurry was diluted to 0.5% consistency and then the dispersed ink particles were removed out of the pulp slurry using the laboratory flotation cell in the same way described in Example 1. In this case, to increase the ink removal efficiency and selectivity, a small amount of the conventional fatty acid collector, SERFAX MT-90, 0.3% based on dry weight of wastepaper, was added prior to the air flotation step and the flotation time was 3 minutes.

**Comparative Example**

To compare to the enzyme de-inked pulp, the conventional de-inked pulp was prepared by the same way, but the following chemicals and conditions were used:

1% NaOH on dry weight of wastepaper
0.1% IGEPAL-660 dispersant
0.8% SERFAX MT-90
50°C pulping temperature
30 minutes pulping time
220 ppm calcium salt addition to the flotation cell
3 minutes flotation time

The brightness and the strength properties of the resulting pulp samples were compared in Table 3.

| Table 3 | Comparison of pulp properties recycled by the method of the present invention and the conventional method. | | |
|---|---|---|---|
| | Brightness (%) | Dirt Amount (count/area) | Tensile Index (N.m/g) |
| enzyme + MT-90 (0.3%) | 79.0 | 450 | 34.3 |
| conventional method + MT-90 (0.8%) | 80.6 | 4,330 | 26.3 |

**De-inking of Waste Newsprint by Pectinolytic Enzyme.**

**Example 3**

As per the method of Example 1, the waste newsprint containing 0.1% of pectinase was soaked for 10 minutes at 40°C and disintegrated for 5 minutes. The disintegrated pulp was diluted to a consistency of 1%. Ink particles were removed by flotation for 1 minute.

**Comparative Example**

The procedure of the comparative example of Example 1 above was repeated.

As shown in Table 4, the brightness and the tensile strength of paper sheet prepared by the method of the present invention are improved.

| Table 4 | Comparison of the method of using pectinolytic enzyme with the conventional method | |
|---|---|---|
| | Brightness (%) | Tensile Index (N.m/g) |
| pectinase enzyme | 44.2 | 33.3 |
| conventional method MT-90 (0.8%) | 38.4 | 32.8 |

The foregoing disclosure and description of the invention is illustrative and explanatory thereof, and various changes in the method steps may be made within the scope of the appended claims without departing from the spirit of the invention.

What Is Claimed Is:

1. A method of de-inking waste printed paper, comprising:

pulping and de-inking waste printed paper in an aqueous medium consisting of water and an enzyme de-inking agent capable of dislodging and dispersing ink particles from pulped paper; and

removing the dislodged and dispersed ink particles from the resulting pulp-containing medium.

2. The method of Claim 1 wherein said ink particles are removed by flotation.

3. The method of Claim 1 wherein said ink particles are removed by washing.

4. The method of Claim 1 wherein the amount of enzyme used is in the range of about 0.005 to about 5.0 percent-by-weight based on the dry weight of the wastepaper.

5. The method of Claim 1 wherein said enzyme is selected from the class consisting of cellulase, hemicellulase, pectinase, other carbohydrases and mixtures thereof.

6. The method of Claim 4 wherein said enzyme is a cellulase selected from the group consisting of cellulases derived from *Trichoderma viride*, *Aspergillus niger* and mixtures thereof.

7. The method of Claim 5 wherein the amount of enzyme used is in the range of about 0.005 to about 5.0 percent-by-weight based on the dry weight of the wastepaper.

8. The method of Claim 1 including controlling the pH of said aqueous medium from about 3 to about 7 and controlling the temperature of the pulping process in a range of from about 20°C up to about 60°C.

9. A method of de-inking waste printed paper, comprising:

converting waste printed paper to a pulp by contacting said paper with an aqueous medium having a pH less than about 7 and comprising water and a de-inking agent consisting of an enzyme;

dislodging ink particles from said waste printed paper by the activity of said de-inking agent; and

removing said dislodged ink particles from the resulting pulp-containing medium.

10. The method of Claim 9 wherein said enzyme is used in an amount between about 0.005 and about 5.0 percent-by-weight based on the dry weight of said wastepaper.

11. The method of Claim 9 including controlling the pH of said pulping and dislodging processes between about 3 and about 7.

12. The method of Claim 9 including controlling the temperature of said pulping and dislodging processes between about 20°C and about 60°C.

13. The method of Claim 9 wherein said enzyme is a carbohydrase.

14. The method of Claim 13 wherein said carbohydrase is selected from the group consisting of cellulase, hemicellulase, pectinase and mixtures thereof.

15. The method of Claim 13 wherein said carbohydrase is a cellulase selected from the group consisting of cellulases derived from *Trichoderma viride, Aspergillus niger* and mixtures thereof.

16. The method of Claim 13 wherein said carbohydrase is used in an amount between about 0.005 and about 5.0 percent-by-weight based on the dry weight of said waste paper.

17. The method of Claim 13 including controlling the pH between about 3 and about 7 and controlling the temperature between about 20 °C and about 60°C during said pulping and dislodging processes.

18. The method of Claim 9 wherein said aqueous medium consists of water and said enzyme.

19. A method of de-inking waste printed paper, comprising:

converting waste printed paper to a pulp in an aqueous medium comprising water and a de-inking agent consisting of an enzyme selected from the group consisting of cellulase, hemicellulase, pectinase, other carbohydrases and mixtures thereof wherein said enzyme is used in an amount between about 0.005 and about 5.0 percent-by-weight based on the dry weight of said waste printed paper, said contacting being carried out in absence of

any chemical de-inking agent at a temperature between about 20°C and about 60°C and at a pH between about 3 and about 7; and

removing dislodged ink particles from the resulting pulp-containing medium by washing or flotation.

20. The method of Claim 19 wherein said enzyme is a cellulase selected from the group of cellulases derived from *Trichoderma viride*, *Aspergillus niger* or mixtures thereof.

## Abstract of the Disclosure

A method of de-inking wastepaper by pulping the paper in the presence of an enzyme is disclosed. The enzyme dislodges the ink particles from the paper fibers. The preferred enzymes are the acid resistant carbohydrases. The de-inking medium is an aqueous solution of the enzyme, preferably maintained at a pH less than about 7 and at a temperature between about 20°C and about 60°C. No conventional chemical de-inking agents are required. The dislodged ink particles may be removed by any conventional method, such as flotation or washing.

# COMBINED DECLARATION AND POWER OF ATTORNEY

*(ORIGINAL, DESIGN, NATIONAL STAGE OF PCT, SUPPLEMENTAL, DIVISIONAL, CONTINUATION OR CIP)*

As a below named inventor, I hereby declare that:

## TYPE OF DECLARATION

This declaration is of the following type: (*check one applicable item below)*

( ) original
( ) design
( ) supplemental

*NOTE: If the declaration is for an International Application being filed as a divisional, continuation or continuation-in-part application do not check next item; check appropriate one of last three items.*

( ) national stage of PCT

*NOTE: If one of the following 3 items apply then complete and also attach ADDED PAGES FOR DIVISIONAL, CONTINUATION OR CIP.*

( ) divisional
( ) continuation
(x) continuation-in-part (CIP)

## INVENTORSHIP IDENTIFICATION

WARNING: *If the inventors are each not the inventors of all the claims an explanation of the facts, including the ownership of all the claims at the time the last claimed invention was made, should be submitted.*

My residence, post office address and citizenship are as stated below next to my name, I believe I am the original, first and sole inventor (*if only one name is listed below)* or an original, first and joint inventor (*if plural names are listed below*) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

## TITLE OF INVENTION

BIOLOGICAL DE-INKING METHOD

## SPECIFICATION IDENTIFICATION

the specification of which: (*complete (a), (b) or (c)*)

(a) ( ) is attached hereto.

(b) (x) was filed on May 6, 1994 as (x) Serial No. 08/239,313 or ( ) Express Mail No._____, *as Serial No. not yet known* and was amended on _____ (*if applicable*).

*NOTE: Amendments filed after the original papers are deposited with the PTO which contain new matter are not accorded a filing date by being referred to in the declaration. Accordingly, the amendments involved are those filed with the application papers or, in the case of a supplemental declaration, are those amendments claiming matter not encompassed in the original statement of invention or claims. See 37 C.F.R.*

(c) ( ) was described and claimed in PCT International Application No. _____ filed on _____ and as amended under PCT Article 19 on _____ (*if any*).

## ACKNOWLEDGEMENT OF REVIEW OF PAPERS AND DUTY OF CANDOR

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to the examination of this application in accordance with Title 37, Code of Federal Regulations. §1.56(a).

( ) In compliance with this duty there is attached an information disclosure statement. 37 CFR §1.97.

## PRIORITY CLAIM

I hereby claim foreign priority benefits under Title 35, United States Code, §119 of any foreign application(s) for patent or inventor's certificate or of any PCT international application(s) designating at least one country other than the United States of America listed below and have also identified below any foreign application(s) for patent or inventor's certificate or any PCT international application(s) designating at least one country other than the United States of America filed by me on the same subject matter having a filing date before that of the application(s) of which priority is claimed.

*(complete (d) or (e))*

(d) ( ) no such applications have been filed.

(e) (x) such applications have been filed as follows.

*NOTE: Where item (c) is entered above and the International Application which designated the U.S. claimed priority check item (e), enter the details below and make the priority claim.*

EARLIEST FOREIGN APPLICATION(S), IF ANY FILED WITHIN 12 MONTHS (6 MONTHS FOR DESIGN) PRIOR TO THIS U.S. APPLICATION

| COUNTRY | APPLICATION NUMBER | DATE OF FILING (day, month, year) | PRIORITY CLAIMED UNDER 35 USC 119 |
|---|---|---|---|
| KOREA | 6514/1989 | 16 MAY 1989 | YES |
| | | | |
| | | | |
| | | | |
| | | | |

ALL FOREIGN APPLICATION(S), IF ANY FILED MORE THAN 12 MONTHS (6 MONTHS FOR DESIGN) PRIOR TO THIS U.S. APPLICATION

_____

_____

_____

## POWER OF ATTORNEY

As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith. (*List name and registration number*)

Margaret E. Anderson, Reg. No. 26,828; Walter R. Brookhart, Reg. No. 29,518; C. James Bushman, Reg. No. 24,810; Loren G. Helmreich, Reg. No. 29,389; William E. Johnson, Jr., Reg. No. 22,719; and Kenneth L. Nash, Reg. No. 34,399

( ) Attached as part of this declaration and power of attorney is the authorization of the above-named attorney(s) to accept and follow instructions from my representative(s).

---

SEND CORRESPONDENCE TO

Mr. Walter R. Brookhart
Browning Bushman, Anderson & Brookhart
5718 Westheimer, Suite 1800
Houston, TX 77057

DIRECT TELEPHONE CALLS TO:
*(Name and telephone number)*

Mr. Walter R. Brookhart
(713) 266-5593

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## DECLARATION

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

### SIGNATURE(S)

Full name of sole or first joint inventor, if any:

Inventor's signature: Steven Say-kyoun OW

Date: June 14, 1996 Country of Citizenship: U.S.A.

Residence: Daejonjikhal-shi, Korea

Post office Address: 21-142, Samboo Apartment, #407 Taepung-dong

Joong-ky, Daejonjikhal-shi, Korea

Full name of second inventor: Tae Jin, Eom

Inventor's signature: omitted inventor - completed on added page

Date: _____ Country of Citizenship: Korea Residence:

Daejonjikhal-shi, Korea

Post office Address: 3-608 Dongsan Apartment Galma-dong Seo-ku,

Daejonjikhal-shi, Korea

*CHECK PROPER BOX(ES) FOR ANY OF THE FOLLOWING ADDED PAGE(S) WHICH FORM A PART OF THIS DECLARATION*

( ) Signature for third and subsequent joint inventors. Number of pages added ____.

( ) Signature by administrator(trix), executor(trix) or legal representative for deceased or incapacitated inventor. Number of pages added _____.

(x) Signature for inventor who refuses to sign or cannot be reached by person authorized under 37 CFR §1.47. Number of pages added __1__.

* * *

(x) Added pages to combined declaration and power of attorney for divisional, continuation, or continuation-in-part (CIP) application.

(x ) Number of pages added __2__

* * *

( ) Authorization of attorney(s) to accept and follow instructions from representative.

* * *

If no further pages form a part of this Declaration then end this Declaration with this page and check the following item.

( ) This declaration ends with this page

## ADDED PAGE TO COMBINED DECLARATION AND POWER OF ATTORNEY FOR SIGNATURE BY ONE JOINT INVENTOR ON BEHALF OF OMITTED INVENTOR(S) WHO REFUSES TO SIGN OR CANNOT BE REACHED (37 CFR 1.47(a))

*Note:* *Any available joint inventor can sign the declaration on behalf of one or more joint inventors who refuse to sign or cannot be reached. Use separate added page for each omitted inventor.*

I. I am an above named joint inventor and have signed this declaration on my own behalf and also sign this declaration under 37 CFR 1.47(a) on behalf of the omitted joint inventor, particulars for whom are:

Full name of (first, second, etc.) Tae Jin, Eom omitted inventor who

____ refuses to sign

__X__ cannot be found or reached

*NOTE:* *The name of the omitted inventor(s) should preferably also be filled in at the appropriate prior space in the declaration adding the words "omitted inventor - completed on added page."*

Republic of Korea
*Country of Citizenship of omitted inventor*

5-211, Daejayen Icha Apartment, Pa-dong, Suseong-ku, Taegu-shi, Republic of Korea
*Last known address of omitted inventor*

*NOTE:* *Ordinarily, the last known address will be the last known residence of the omitted inventor(s). Other addresses at which the omitted inventor(s) may be reached should also be given. These can best be given in the Declaration Of Facts In Support Of Filing On Behalf Of Omitted Inventor. MPEP § 4.09.03(e).*

II. Accompanying this declaration is:

(1) A DECLARATION OF FACTS IN SUPPORT OF FILING ON BEHALF OF OMITTED INVENTOR

(2) THE PETITION FEE OF $130.00 (37 CFR 1.17(h))

Steven Say-Kyoun OW
*(type or print name of joint inventor signing on behalf of omitted inventor)*

Date: June 14 1994

*(Signature of joint inventor signing on behalf of omitted inventor)*

PRINT OF DRAWINGS
AS ORIGINALLY FILED

# United States Patent & Trademark Office

Office of Initial Patent Examination -- Scanning Division

## Application deficiencies found during scanning:

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